



UNIVERSITAT DE BARCELONA



Divisió de Ciències Experimentals
i Matemàtiques
Facultat de Química
Departament d'Enginyeria Química
i Metal·lúrgia

Ferran Espiell
Catedràtic

Martí i Franquès, 1
08028 Barcelona
Tel. 93 402 13 16
Fax 93 402 12 91

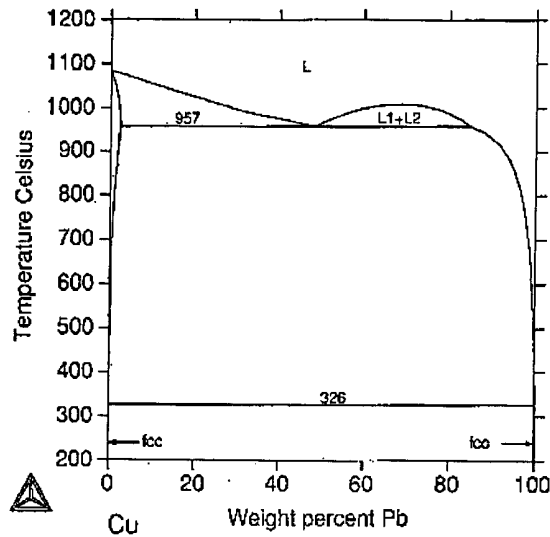
DECLARATION OF Dr. FERRAN ESPIELL

Hon. Commissioner of
Patents and Trademarks Office
Washington DC 20231

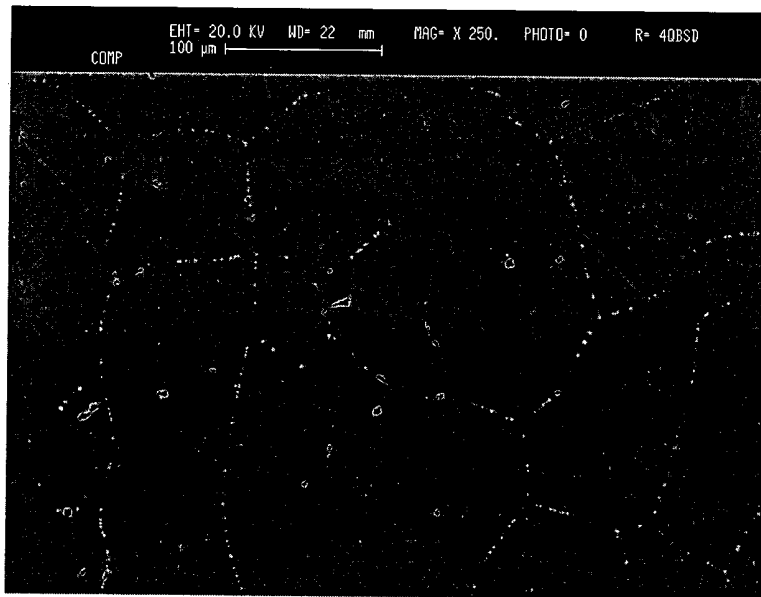
Sir:

I, Dr Ferran Espiell, declare that

1. I am over 21 years old
2. I am co-inventor of the U.S. patent application Serial No: 09/499,207
3. I received a Ph.D. from the University of Barcelona and currently am a full professor at the University of Barcelona.
4. Lead is the only element that permits to produce copper by continuous casting. We have tested all the other possible elements with compositions as those described in the patent, but none of them give positive results.
5. Concerning the content of 200 ppm of lead: the influence of lead content on the copper casting is tested by increasing the amount of lead in the liquid copper until it can be cast without breaking. In fact, this test is performed every day by the manufacturer, in each casting, and we have corroborated that a lead content above 200 ppm is required to make possible the continuous casting, as is summarized in my declaration on April 2003.
6. Lead forms insoluble inclusions in solid copper that dissipate tensions among grains thus avoiding bar break. The following figure shows the copper-lead phase diagram, where the monotectic reaction can be observed at high copper contents for Cu-Pb alloys.



7. The following picture shows a micrograph obtained by Scanning Electron Microscopy with Back Scattering Electrons image where lead inclusions can be observed as the lighter zones (heavier mean atomic weight) in the grain limits. This micrograph corresponds to sample 3 described in my previous declaration dated April 2003.



8. The composition we claim comply with ASTM standards B224-98 "Standard classification of copper" and B49-98 "Standard specification for Copper Rod Drawing Stock for Electrical Purposes" that demand for this application an electrical conductivity of 100% IACS minimum and a copper content 99.90% minimum. We enclose a hard copy of this standard.

9. The Japanese patent No 62133050 claims for a Cu alloy containing at least 0.1% of Ti and 0.1% of Sn as main alloying elements. With these contents in Ti and Sn this alloy do not meet the requirements of Copper for electrical purposes, either in Cu content (they will have less than 99.9% copper) and electrical conductivity. Moreover, with these Ti and Sn contents electrical conductivity can not achieve in any case values close to 100% IACS.
10. The Japanese patent No 62133050 refers to a Cu alloy obtained after consecutive heat treatments and gives as an example a final product having a tensile strength of 120 kg/mm² and an electrical conductivity of only 30% IACS suitable for precision springs, but not for electrical purposes.
11. Concerning copper alloys containing both titanium and tin, we have not found any reference in the literature to such alloys with Ti and Sn contents in the range of that cited in the patent, that present conductivities of 100% IACS, not even above 70% IACS. As examples we can mention a copper-tin alloy with 1.0-1.7% tin (C50500 alloy) that presents a conductivity of 48% IACS (Metals Handbook, vol. 2, 10th ed., ASM International, USA, 1990), or a copper alloy with 1.5% titanium, with a conductivity of 25% IACS (S. Nagarjuna and DS. Sarma, Titanium (India), vol.5, n.4, pp.16-38, Nov. 2000). These alloys are not usually used as high conductivity copper.

I hereby declare that all statements of my own knowledge are true and that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that said wilful false statements may jeopardize the validity of the application or any patent issued therein.

Dated: September 25th, 2003

Dr. Ferran Espiell

A handwritten signature in black ink, appearing to read 'F. Espiell', written over a horizontal line.

EXPRESS MAIL CERTIFICATE

Date

I hereby certify that, on the date indicated above, the paper or fee was deposited with the U.S. Postal Service & that it was addressed for delivery to the Assistant Commissioner for Patents, Washington, DC 20231 by "Express Mail Post Office to Addressee" service.

Name (Print)

Signature

Customer No.:



07278

PATENT TRADEMARK OFFICE

PLEASE CHARGE ANY DEFICIENCY UP TO \$300.00 OR CREDIT ANY EXCESS IN THE FEES DUE WITH THIS DOCUMENT TO OUR DEPOSIT ACCOUNT NO. 04 - 0100

RECEIVED
OCT 14 2003
GROUP 1700

Docket No: 2136/00684

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Jose GUIXA ARDERIU et al.

Serial No.: 09/499,207

Art Unit: 1742

Confirmation No.: 9917

Filed: February 7, 2000

Examiner: S. Ip

For: MANUFACTURE OF COPPER MICROALLOYS

DECLARATION OF DR. FERRAN ESPIELL

Hon. Commissioner of
Patents and Trademarks
Washington, DC 20231

Sir:

I, Dr. Ferran Espiell, declare that:

1. I am over 21 years of age.
2. I am a co-inventor of the above-identified U.S. patent application.
3. I received a Ph.D. from the University of Barcelona and currently am a full professor at the University of Barcelona.

4. The following experiment was conducted under my supervision and control.

5. The three copper microalloys having the following compositions were prepared:

Element	Sample 1 (ppm of element)	Sample 2 (ppm of element)	Sample 3 (ppm of element)
Zn	22.5	22.5	25.0
Pb	196.0	182.0	208.0
Sn	103.9	103.0	92.5
P	<0.8	<0.8	0.7
Mn	2.1	2.1	1.4
Fe	13.3	13.3	17.0
Ni	69.3	69.3	83.3
Si	7.2	7.2	6.5
Mg	-	0.0	0.6
Cr	0.1	0.1	0.4
Te	11.6	11.6	9.0
As	14.4	14.4	19.3
Se	5.5	6.5	10.1
Sb	14.3	14.3	11.8
Cd	3.0	3.0	2.1
Bi	5.0	5.0	4.8

Ag	141.0	141.0	121.9
Co	0.8	0.8	0.4
Al	1.3	1.3	0.9
S	13.9	13.9	17.7
Cu	99.94	99.94	99.94

6. Electrolytically refined copper cathode and copper scrap were melted at 1150° C. A fluxing agent was introduced at a rate of 8 kg per ton of scrap. The reaction was allowed to proceed for 30 minutes and a slag was then removed.

7. After removal of the slag, air was injected into the furnace to increase the oxygen concentration up to 2500 grams per ton. A second fluxing material was introduced and after about 30 minutes, a second slag was removed. After removed the second slag, As, Sb, and Sn were removed by increasing the concentration of oxygen in the molten material up to 6000-8000 grams per ton (ppm) and introducing 10 Kg of a third fluxing agent per ton of copper. After 30 minutes the contents of the elements were tested to determine whether they were within previously established margins. Once positive, the oxygen content in the molten product was reduced to 200 grams per ton (ppm). The metal was analyzed and, where necessary, lead and other metals were added to obtain the desired concentrations.

8. The copper microalloy was then cast by adjusting the temperature to 1150° C, casting the copper microalloy according to the "Continuous Properi" system with a casting speed of 12-15 tons per hour, and solidifying and rolling to obtain an 8 mm rod.

9. Samples 1 and 2 showed cracks and breakages in the rods cast, thereby preventing them from being rolled. In contrast, Sample 3 did not exhibit cracks and breakages and therefore could be rolled. Copper microalloy is typically sold as a rolled product.

I hereby declare that all statements of my own knowledge are true and that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that said willful false statements may jeopardize the validity of the application or any patent issued therein.

Dated: April, 1st, 2003


Dr. Ferran Espiell